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US DEPARTMENT OF COMMERCE PATENT AND TRADEMARK OFFICE

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2001-1443ATRANSMITTAL LETTER TO THE UNITED STATES  
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CONCERNING A FILING UNDER 35 U.S.C. §371U.S. APPLICATION NO.  
(if known, enter in Class 3)  
[NEW] 09/937724International Application No.  
PCT/EP00/02552International Filing Date  
March 23, 2000Priority Date Claimed  
April 13, 1999**Title of Invention**

SINGLET OXYGEN OXIDATION OF ORGANIC SUBSTRATES

**Applicant(s) For DO/EO/US**


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Applicant herewith submits to the United States Designated/Elected Office (DO/EO/US) the following items and other information:

1. ☒ This is a **FIRST** submission of items concerning a filing under 35 U.S.C. §371.
2. ☐ This is a **SECOND** or **SUBSEQUENT** submission of items concerning a filing under 35 U.S.C. §371.
3. ☐ This express request to begin national examination procedures (35 U.S.C. §371(f)) at any time rather than delay examination until the expiration of the applicable time limit set in 35 U.S.C. §371(b) and PCT Articles 22 and 39(1).
4. ☒ A proper Demand for International Preliminary Examination was made by the 19th month from the earliest claimed priority date.
5. ☒ A copy of the International Application as filed (35 U.S.C. §371(c)(2))
  - a. ☐ is transmitted herewith (required only if not transmitted by the International Bureau).
  - b. ☒ has been transmitted by the International Bureau.
  - c. ☐ is not required, as the application was filed in the United States Receiving Office (RO/US)
6. ☒ A translation of the International Application into English (35 U.S.C. §371(c)(2)). **ATTACHMENT A**
7. ☐ Amendments to the claims of the International Application under PCT Article 19 (35 U.S.C. §371(c)(3)).
  - a. ☐ are transmitted herewith (required only if not transmitted by the International Bureau).
  - b. ☐ have been transmitted by the International Bureau.
  - c. ☐ have not been made; however, the time limit for making such amendments has NOT expired.
  - d. ☐ have not been made and will not be made.
8. ☐ A translation of the amendments to the claims under PCT Article 19.
9. ☒ An oath or declaration of the inventor(s) (35 U.S.C. §371(c)(4)). **ATTACHMENT B**
10. ☐ A translation of the annexes to the International Preliminary Examination Report under PCT Article 36 (35 U.S.C. §371(c)(5)).

Items 11. to 14. below concern other document(s) or information included:

11. ☒ An Information Disclosure Statement under 37 CFR 1.97 and 1.98. **ATTACHMENT C**
12. ☒ An assignment document for recording. A separate cover sheet in compliance with 37 CFR 3.28 and 3.31 is included.  
**ATTACHMENT D**
13. ☐ A **FIRST** preliminary amendment.  
☐ A **SECOND** or **SUBSEQUENT** preliminary amendment.
14. ☒ Other items or information:
  - a. Cover Page of Published International Application No. WO 00/61524 - **ATTACHMENT E**
  - b. International Search Report - **ATTACHMENT F**

U.S. APPLICATION NO. [NEW] <b>09/937724</b>		INTERNATIONAL APPLICATION NO. PCT/EP00/02552		ATTORNEY'S DOCKET NO. 2001-1443A	
15. <input checked="" type="checkbox"/> The following fees are submitted  <b>BASIC NATIONAL FEE (37 CFR 1.492(a)(1)-(5)):</b> Neither international preliminary examination fee nor international search fee paid to USPTO and International Search Report not prepared by the EPO or JPO ..... \$1000.00 International Search Report has been prepared by the EPO or JPO ..... \$ 860.00 International preliminary examination fee not paid of USPTO but international search paid to USPTO ..... \$ 710.00 International preliminary examination fee paid to USPTO but claims did not satisfy provisions of PCT Article 33(1)-(4) ..... \$ 690.00 International preliminary examination fee paid of USPTO and all claims satisfied provisions of PCT Article 33(1)-(4) ..... \$ 100.00  <b>ENTER APPROPRIATE BASIC FEE AMOUNT =</b>				CALCULATIONS	PTO USE ONLY
Surcharge of \$130.00 for furnishing the oath or declaration later than [ ] 20 [ ] 30 months from the earliest claimed priority date (37 CFR 1.492(e)).					
Claims	Number Filed	Number Extra	Rate		
Total Claims	8 -20 =	0	X \$18.00		
Independent Claims	1 - 3 =	0	X \$80.00		
Multiple dependent claim(s) (if applicable)			+ \$270.00		
<b>TOTAL OF ABOVE CALCULATIONS =</b>				\$860.00	
[ ] Small Entity Status is hereby asserted. Above fees are reduced by 1/2.					
<b>SUBTOTAL =</b>				\$860.00	
Processing fee of \$130.00 for furnishing the English translation later than [ ] 20 [ ] 30 months from the earliest claimed priority date (37 CFR 1.492(f)).				+	
<b>TOTAL NATIONAL FEE =</b>				\$860.00	
Fee for recording the enclosed assignment (37 CFR 1.21(h)). The assignment must be accompanied by an appropriate cover sheet (37 CFR 3.28, 3.31). \$40 per property +				\$ 40.00	
<b>TOTAL FEES ENCLOSED =</b>				\$900.00	
				Amount to be refunded	\$
				Amount to be charged	\$
a. <input checked="" type="checkbox"/> A check in the amount of \$ <u>900.00</u> to cover the above fees is enclosed. A duplicate copy of this form is enclosed. b. [ ] Please charge my Deposit Account No. 23-0975 in the amount of \$ _____ to cover the above fees. A duplicate copy of this sheet is enclosed. c. <input checked="" type="checkbox"/> The Commissioner is hereby authorized to charge any additional fees which may be required, or credit any overpayment to Deposit Account No. <u>23-0975</u> .					
<b>NOTE: Where an appropriate time limit under 37 CFR 1.494 or 1.495 has not been met, a petition to revive (37 CFR 1.137(a) or (b)) must be filed and granted to restore the application to pending status.</b>					
19. CORRESPONDENCE ADDRESS  <div style="text-align: center;">   <b>000513</b>          PATENT TRADEMARK OFFICE       </div>			By: <u>Matthew Jacob</u> Matthew Jacob Registration No. 25,154  WENDEROTH, LIND & PONACK, L.L.P. 2033 "K" Street, N.W., Suite 800 Washington, D.C. 20006-1021 Phone: (202) 721-8200 Fax: (202) 721-8250  September 28, 2001		

THE COMMISSIONER IS AUTHORIZED  
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FEE FOR THIS PAPER TO DEPOSIT  
ACCOUNT NO. 23-0975.

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[2001-1443A]

Singlet oxygen oxidation of organic substances [sic]

5 The only singlet oxygen oxidation ( $^1\text{O}_2\text{-Ox}$ ) which is currently carried out industrially is the photochemical  $^1\text{O}_2\text{-Ox}$  in which the  $^1\text{O}_2$  is generated by a photochemical route. The disadvantage of this process is given by the high costs of the photochemical equipment required, and by a limited service life. The required lamps degenerate relatively rapidly during the oxidation as a result of soiling of the glass surface. In addition, this process is not suitable for colored substrates. The process is actually suitable only for fine chemicals which are prepared on a relatively small scale. (La Chimica e l'Industria, 1982, Vol. 64, page 156).

15 For this reason, attempts have been made to find other process variants for the  $^1\text{O}_2\text{-Ox}$  which are suitable for the  $^1\text{O}_2\text{-Ox}$  of non-water-soluble, hydrophobic organic substrates.

20 J. Am. Chem. Soc., 1968, 90, 975 describes, for example, the classical "dark"  $^1\text{O}_2\text{-Ox}$  in which  $^1\text{O}_2$  is not generated photochemically, but chemically. In this process, hydrophobic substrates are oxidized by means of a hypochlorite/ $\text{H}_2\text{O}_2$  system in a solvent mixture of water and organic solvent. However, this process has only found a few synthetic applications since many substrates are only sparingly soluble in the required medium. In addition, the use possibility is rather limited because of secondary reactions between hypochlorite and substrate or solvent. In addition, a large part of the  $^1\text{O}_2$  is deactivated in the gas phase. In addition, this process is not suitable for industrial scale since in the organic medium addition of the hypochlorite onto  $\text{H}_2\text{O}_2$  results, and a large excess of  $\text{H}_2\text{O}_2$  is required to suppress the secondary reaction of substrate with hypochlorite. An additional disadvantage arises as a result of the formation of stoichiometric amounts of salt.

J. Am. Chem. Soc., 1997, 119, 5286 and EP-A-0 288 337 describes a process that permits the generation, catalyzed by molybdate in aqueous solution, of  $^1\text{O}_2$  from  $\text{H}_2\text{O}_2$  and is nevertheless suitable for hydrophobic substrates from a relatively wide molecular weight range. In this process, a microemulsion is used as reaction medium. However, use on an industrial scale is associated with problems since product isolation from the microemulsion is difficult. In addition, it is a relatively expensive process since rather large amounts of surfactant have to be used relative to the substrate.

Accordingly, it was an object of the present invention to find an improved method of the "dark"  $^1\text{O}_2$ -Ox which can be used simply, cost-effectively and in an environmentally friendly manner on an industrial scale and is suitable for a large number of substrates.

Unexpectedly, it has now been found that the "dark"  $^1\text{O}_2$ -Ox can be carried out in an extremely efficient manner with high yield in certain organic solvents as reaction medium, without the addition of water as cosolvent and without surfactant.

Accordingly, the present invention provides a process for the oxidation of organic substrates by means of  $^1\text{O}_2$ , which comprises adding 30-70% strength  $\text{H}_2\text{O}_2$  to hydrophobic organic substrates which react with  $^1\text{O}_2$  in an organic solvent in the presence of a heterogeneous or homogeneous catalyst, whereupon, following the catalytic decomposition of  $\text{H}_2\text{O}_2$  to give water and  $^1\text{O}_2$ , oxidation to give the corresponding oxidation products takes place.

The process according to the invention is suitable for the oxidation of hydrophobic organic substrates which react with  $^1\text{O}_2$ .

Accordingly, substrates which may be used are the following compounds: olefins which contain one or more, i.e. up to 10, preferably up to 6, particularly preferably up to 4, C=C double bonds; electron-rich aromatics, such as  $\text{C}_6$ - $\text{C}_{50}$ , preferably up to  $\text{C}_{30}$ , particularly preferably up to  $\text{C}_{20}$ , phenols, polyalkylbenzenes, polyalkoxybenzenes; polycyclic aromatics having 2 to 10, preferably up to 6, particularly preferably up to 4 aromatic rings; sulfides, such as, for example, alkyl sulfides, alkenyl sulfides, aryl sulfides which are either mono- or disubstituted on the sulfur atom, and heterocycles having an O, N or S atom in the ring, such as, for example,  $\text{C}_4$ - $\text{C}_{50}$ , preferably up to  $\text{C}_{30}$ , particularly

preferably up to C<sub>20</sub>, furans, C<sub>4</sub>-C<sub>50</sub>, preferably up to C<sub>30</sub>, particularly preferably up to C<sub>20</sub>, pyrroles, C<sub>4</sub>-C<sub>60</sub>, preferably up to C<sub>30</sub>, particularly preferably up to C<sub>20</sub>, thiophenes. In this connection, the substrates may have one or more substituents, such as halogen (F, Cl, Br, I), cyanide, carbonyl groups, hydroxyl groups, C<sub>1</sub>-C<sub>50</sub>, preferably up to C<sub>30</sub>, particularly preferably up to C<sub>20</sub>, alkoxy groups, C<sub>1</sub>-C<sub>50</sub>, preferably up to C<sub>30</sub>, particularly preferably up to C<sub>20</sub>, alkyl groups, C<sub>6</sub>-C<sub>50</sub>, preferably up to C<sub>30</sub>, particularly preferably up to C<sub>20</sub>, aryl groups, C<sub>2</sub>-C<sub>50</sub>, preferably up to C<sub>30</sub>, particularly preferably up to C<sub>20</sub>, alkenyl groups, C<sub>2</sub>-C<sub>50</sub>, preferably up to C<sub>30</sub>, particularly preferably up to C<sub>20</sub>, alkynyl groups, carboxylic acid groups, ester groups, amide groups, amino groups, nitro groups, silyl groups, silyloxy groups, sulfone groups, sulfoxide groups. In addition, the substrates may be substituted by one or more NR<sup>1</sup>R<sup>2</sup> radicals in which R<sub>1</sub> [sic] or R<sub>2</sub> [sic] may be identical or different and are H; C<sub>1</sub>-C<sub>50</sub>, preferably up to C<sub>30</sub>, particularly preferably up to C<sub>20</sub>, alkyl; formyl; C<sub>2</sub>-C<sub>50</sub>, preferably up to C<sub>30</sub>, particularly preferably up to C<sub>20</sub>, acyl; C<sub>7</sub>-C<sub>50</sub>, preferably up to C<sub>30</sub>, particularly preferably up to C<sub>20</sub>, benzoyl, where R<sup>1</sup> and R<sup>2</sup> may also together form a ring, such as, for example, in a phthalimido group.

Examples of suitable substrates are: 2-butene; isobutene; 2-methyl-1-butene; 2-hexene; 1,3-butadiene; 2,3-dimethylbutene; D<sup>9,10</sup>-octalin [sic], 2-phthalimido-4-methyl-3-pentene; 2,3-dimethyl-1,3-butadiene; 2,4-hexadiene; 2-chloro-4-methyl-3-pentene; 2-bromo-4-methyl-3-pentene; 1-trimethylsilylcyclohexene; 2,3-dimethyl-2-butenyl-para-tolylsulfone; 2,3-dimethyl-2-butenyl-para-tolyl sulfoxide; N-cyclohexenylmorpholine; 2-methyl-2-norbornene; terpinolene; α-pinene; β-pinene; β-citronellol; ocimene; citronellol; geraniol; farnesol; terpinene; limonene; trans-2,3-dimethylacrylic acid; α-terpinene; isoprene; cyclopentadiene; 1,4-diphenylbutadiene; 2-ethoxybutadiene;

1,1'-dicyclohexenyl; cholesterol; ergosterol acetate;  
5-chloro-1,3-cyclohexadiene; 3-methyl-2-buten-1-ol;  
3,5,5-trimethylcyclohex-2-en-1-ol; phenol,  
1,2,4-trimethoxybenzene, 2,3,6-trimethylphenol,  
5 2,4,6-trimethylphenol, 1,4-dimethylnaphthalene, furan,  
furfuryl alcohol, furfural, 2,5-dimethylfuran,  
isobenzofuran, dibenzyl sulfide, (2-methyl-5-tert-  
butyl)phenyl sulfide etc.

10 As a result of the oxidation according to the  
invention, the corresponding oxidation product is  
obtained from the substrates. Alkenes, (polycyclic)  
aromatics or heteroaromatics give, in particular,  
hydroperoxides or peroxides which are able to further  
15 react under the reaction conditions to give alcohols,  
epoxides, acetals or carbonyl compounds, such as  
ketones, aldehydes, carboxylic acids or esters, if the  
hydroperoxide or the peroxide is unstable.

20 The oxidation according to the invention is carried out  
in an organic solvent.

Suitable solvents are C<sub>1</sub>-C<sub>8</sub>-alcohols, such as methanol,  
ethanol, propanol, isopropanol, butanol, isobutanol;  
ethylene glycol, propylene glycol, formamide,  
25 N-methylformamide, dimethylformamide, sulfolane,  
propylene carbonate.

Preference is given to using methanol, ethanol,  
propanol, isopropanol, ethylene glycol, propylene  
glycol, formamide, N-methylformamide or  
30 dimethylformamide, particularly preferably methanol,  
ethanol, ethylene glycol, propylene glycol, formamide  
or dimethylformamide as solvent.

Up to 25% of water may optionally be added to the  
organic solvent. However, the addition of water does  
35 not bring any advantages for the reaction. Water is  
therefore preferably not added.

A metal which is suitable for <sup>1</sup>O<sub>2</sub> oxidations and is  
described, for example, in J. Am. Chem. Soc., 1985,

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107, 5844 or in Membrane Lipid Oxid. Vol. II, 1991, 65, is added as heterogeneous or homogeneous inorganic catalyst to the solvent/substrate mixture.

In this connection, the metal can be in forms customary  
5 for  $^1\text{O}_2$  oxidations, for example as the oxide, oxo complex, nitrate, carboxylate, hydroxide, carbonate, chloride, fluoride, sulfate, tetrafluoroborate, etc.

Preference is given to catalysts based on molybdenum, tungsten, scandium, vanadium, titanium, zirconium,  
10 praseodymium, neodymium, samarium, europium, terbium, dysprosium, holmium, erbium, ytterbium and lutetium. Particular preference is given to molybdenum catalysts.

The amount of catalyst used depends on the substrate  
15 used and is between 1 and 50 mol%, preferably between 5 and 25 mol%.

This is followed by the addition of 30-70% strength, preferably 40-60% strength,  $\text{H}_2\text{O}_2$ .  $\text{H}_2\text{O}_2$  is preferably  
20 added slowly or in portions to the reaction mixture of solvent, substrate and catalyst, the reaction mixture being stirred.

In the process according to the invention, the  
25 consumption of  $\text{H}_2\text{O}_2$  is dependent on the substrate used. For reactive substrates, 2 to 3 equivalents of  $\text{H}_2\text{O}_2$  are preferably needed, while less reactive substrates are preferably reacted with 3 to 10 equivalents of  $\text{H}_2\text{O}_2$ .

30 The reaction temperature is between 0 and 50°C, preferably between 15 and 35°C.

In some cases, to improve activation of the catalyst, it may be advantageous to add customary basic or acidic  
35 additives to the reaction mixture.

The course of the reaction can be monitored by means of UV spectroscopy or by means of HPLC. When the reaction is complete, i.e. after 1 to 30 hours, depending on the

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reaction conditions, the reaction mixture is worked up. Removal of the catalyst has proven unexpectedly simple particularly when molybdate catalysts are used, such as e.g. sodium molybdate, in some solvents. Although the  
5 reaction proceeds completely homogeneously when molybdate catalysts, such as, for example,  $\text{Na}_2\text{MoO}_4 \cdot 2\text{H}_2\text{O}$  in mono-hydroxylic, alcoholic solvents, i.e. in monohydric alcohols such as, for example, methanol or ethanol, are used, provided  $\text{H}_2\text{O}_2$  is added, the catalyst,  
10 after all of the  $\text{H}_2\text{O}_2$  has been added, precipitates out of the reaction mixture, as a result of which the catalyst can be separated off by simple centrifugation or filtration, and recycled.

15 The end-product which remains can, where appropriate, be purified by means of recrystallization, extraction or distillation.

The process according to the invention permits the  
20 oxidation of a large number of hydrophobic compounds and is particularly advantageous for the oxidation of water-insoluble substrates which could not be oxidized with hitherto known chemical methods with high yield. Accordingly, the process according to the invention is  
25 particularly suitable for the oxidation of unsaturated organic compounds, such as terpenes, for example  $\alpha$ -terpinene and citronellol, aromatic polycycles, steroids, furans, cyclopentadienes, phenols etc., and generally for all compounds which react with  $^1\text{O}_2$ .

30 The process according to the invention gives the desired end-products in high yields of up to 100% with high purity.

The process according to the invention is characterized  
35 by the simple process regime which is best suited to the industrial scale since it can take place in simple multipurpose plants and with simple work-up steps, and can be used for a wide spectrum of substrates.

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Example 1:

a) 0.2 ml of an aqueous 2 mol/l  $\text{Na}_2\text{MoO}_4$  solution or  
b) 0.4 mmol of  $\text{Na}_2\text{MoO}_4 \cdot 2\text{eq}$ .  
5 were added to a thermostated solution ( $25^\circ\text{C}$ ) of 2 mmol  
of an organic substrate ( $\alpha$ -terpinene or  $\beta$ -citronellol)  
in 4 ml of an organic solvent. 0.08 ml of  $\text{H}_2\text{O}_2$  (50%)  
were added to this mixture. After the reaction mixture  
had turned yellow again, two further 0.08 ml portions  
10 of  $\text{H}_2\text{O}_2$  (50%) were added. The course of the reaction was  
monitored in the case of  $\alpha$ -terpinene by means of UV  
spectroscopy (266 nm) and in the case of  $\beta$ -citronellol  
by means of HPLC (MeOH/ $\text{H}_2\text{O}$  70/30).

15 Example 2:

a) 0.2 ml of an aqueous 2 mol/l  $\text{Na}_2\text{MoO}_4$  solution or  
b) 0.4 mmol of  $\text{Na}_2\text{MoO}_4 \cdot 2\text{eq}$ .  
were added to a thermostated solution ( $25^\circ\text{C}$ ) of 2 mmol  
20 of an organic substrate ( $\alpha$ -terpinene or  $\beta$ -citronellol)  
in 4 ml of an organic solvent. 0.25 ml of  $\text{H}_2\text{O}_2$  (50%)  
were added to this mixture in one portion. The course  
of the reaction was monitored in the case of  $\alpha$ -  
terpinene by means of UV spectroscopy (266 nm) and in  
25 the case of  $\beta$ -citronellol by means of HPLC (MeOH/ $\text{H}_2\text{O}$   
70/30).

The solvents used and the conversion of  $\alpha$ -terpinene  
into ascaridol and  $\beta$ -citronellol into a 1/1 mixture of  
30 the corresponding hydroperoxides are given in table 1:

Table 1:

Example	Substrate	Solvent	Cat. a) or b)	Conversion
1	$\alpha$ -terpinene	Methanol	a	100% after 2 h
1	$\alpha$ -terpinene	Methanol	b	>95% after 2 h
2	$\alpha$ -terpinene	Methanol	a	>95% after 2 h
1*	$\alpha$ -terpinene	Ethanol	a	90% after 22 h
1	$\alpha$ -terpinene	Formamide	a	70% after 21 h
1	$\alpha$ -terpinene	N-Me-formamide	a	75% after 3 h
1	$\alpha$ -terpinene	DMF	a	64% after 21 h
1	$\alpha$ -terpinene	Sulfolane	a	70% after 21 h
1	$\beta$ -citronellol	Methanol	a	80% after 3 h
1	$\beta$ -citronellol	Formamide	a	95% after 3 h

\* 3rd portion of  $H_2O_2$  was 0.09 ml

5

Example 3: Product isolation from reaction mixture with methanol as solvent

10 After the time given in table 1, the precipitated-out catalyst was removed from the reaction mixture by means of centrifugation. The precipitate was washed twice with absolute ethanol, and the combined solvent batches (methanol and ethanol) were removed on a rotary evaporator. The oxidation product which remained was  
15 dissolved in  $CDCl_3$  for NMR analysis. In the case of  $\alpha$ -terpinene, analysis confirmed the formation of a virtually quantitative amount of >95% pure ascaridol. In the case of  $\beta$ -citronellol, approximately 80% of product were obtained which, according to NMR analysis,  
20 consisted of a 1/1 mixture of the two corresponding hydroperoxides.

Example 4:

25 At 35°C, 45  $\mu$ l of  $H_2O_2$  (50%) were added to a solution of 325  $\mu$ l of  $\alpha$ -terpinene and 48.5 mg of  $Na_2MoO_4 \cdot 2H_2O$  in 4 ml of methanol. Five further 45  $\mu$ l portions of  $H_2O_2$

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(50%) were added to this mixture as soon as the red-colored reaction mixture turned yellow again. After 1.5 hours, the reaction mixtures [sic] was analyzed by means of HPLC. Analysis gave a quantitative formation of ascaridol.

Example 5.

At 25°C, 80 µl of H<sub>2</sub>O<sub>2</sub> (50%) were added to a solution of 365 µl of citronellol and 97 mg of Na<sub>2</sub>MoO<sub>4</sub>·2H<sub>2</sub>O in 4 ml of ethylene glycol. After 1, 2 and after 19 hours, 3 further 80 µl portions of H<sub>2</sub>O<sub>2</sub> (50%) were added to this mixture. HPLC analysis gave a 100% conversion with a yield of secondary hydroperoxide of 38% and a yield of tertiary hydroperoxide of 62%.

Patent claims

1. A process for the oxidation of organic substrates by means of  $^1\text{O}_2$ , which comprises adding 30-70% strength  $\text{H}_2\text{O}_2$  to hydrophobic organic substrates which react with  $^1\text{O}_2$  in an organic solvent in the presence of a heterogeneous or homogeneous catalyst, whereupon, following the catalytic decomposition of  $\text{H}_2\text{O}_2$  to give water and  $^1\text{O}_2$ , oxidation to give the corresponding oxidation products takes place.
2. The process as claimed in claim 1, wherein the substrates which react with  $^1\text{O}_2$  used are olefins which contain 1 to 10  $\text{C}=\text{C}$  double bonds;  $\text{C}_6$ - $\text{C}_{50}$  phenols, polyalkylbenzenes, polyalkoxybenzenes; polycyclic aromatics having 2 to 10 aromatic rings; alkyl sulfides, alkenyl sulfides, aryl sulfides which are either mono- or disubstituted on the sulfur atom, and  $\text{C}_4$ - $\text{C}_{50}$  heterocycles having an O, N or S atom in the ring, which may be unsubstituted or may be mono- or polysubstituted by halogens, cyanide, carbonyl groups, hydroxyl groups,  $\text{C}_1$ - $\text{C}_{50}$  alkoxy groups,  $\text{C}_1$ - $\text{C}_{50}$  alkyl groups,  $\text{C}_6$ - $\text{C}_{50}$  aryl groups,  $\text{C}_2$ - $\text{C}_{50}$  alkenyl groups,  $\text{C}_2$ - $\text{C}_{50}$  alkynyl groups, carboxylic acid groups, ester groups, amide groups, amino groups, nitro groups, silyl groups, silyloxy groups, sulfone groups, sulfoxide groups or by one or more  $\text{NR}^1\text{R}^2$  radicals in which  $\text{R}_1$  [sic] or  $\text{R}_2$  [sic] may be identical or different and are H;  $\text{C}_1$ - $\text{C}_{50}$  alkyl; formyl;  $\text{C}_2$ - $\text{C}_{50}$  acyl;  $\text{C}_7$ - $\text{C}_{50}$  benzoyl, where  $\text{R}^1$  and  $\text{R}^2$  may also together form a ring.
3. The process as claimed in claim 1, wherein the solvent used is  $\text{C}_1$ - $\text{C}_8$ -alcohols, formamide, N-methylformamide, dimethylformamide, sulfolane, propylene carbonate.

4. The process as claimed in claim 4, wherein the solvent used is methanol, ethanol, propanol, isopropanol, ethylene glycol, propylene glycol, formamide, N-methylformamide or dimethylformamide.
- 5
5. The process as claimed in claim 1, wherein catalysts based on molybdenum, tungsten, scandium, vanadium, titanium, zirconium, praseodymium, neodymium, samarium, europium, terbium, 10 dysprosium, holmium, erbium, ytterbium or lutetium in the form of oxides, oxo complexes, nitrates, carboxylates, hydroxides, carbonates, chlorides, fluorides, sulfates or tetrafluoroborates are used.
- 15
6. The process as claimed in claim 1, wherein 2 to 10 equivalents of  $H_2O_2$  are used depending on the substrate used.
- 20
7. The process as claimed in claim 1, wherein the reaction temperature is between 0 and 50°C.
8. The process as claimed in claim 1, wherein, following the reaction of the hydrophobic organic substrates which react with  $^1O_2$  in a monohydric  $C_1$ - $C_8$  alcohol as solvent in the presence of a molybdate catalyst with 30-70% strength  $H_2O_2$  to 25 give the corresponding oxidation products, the removal and recycling of the precipitated-out catalyst when the reaction is complete is carried 30 out by simple centrifugation or filtration.

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## DECLARATION AND POWER OF ATTORNEY FOR U.S. PATENT APPLICATION

( ) Original ( ) Supplemental ( ) Substitute (X) PCT ( ) Design

As a below named inventor, I hereby declare that: my residence, post office address and citizenship are as stated below next to my name; that I verily believe that I am the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural inventors are named below) of the subject matter which is claimed and for which a patent is sought on the invention entitled:

Title: \_\_\_\_\_

of which is described and claimed in:

- ( ) the attached specification, or  
 ( ) the specification in the application Serial No. \_\_\_\_\_ filed \_\_\_\_\_;  
 and with amendments through \_\_\_\_\_ (if applicable), or  
 (X) the specification in International Application No. PCT/ EPO0/2552, filed 23/03/2000, and as amended  
 on \_\_\_\_\_ (if applicable).

I hereby state that I have reviewed and understand the content of the above-identified specification, including the claims, as amended by any amendment(s) referred to above.

I acknowledge my duty to disclose to the Patent and Trademark Office all information known to me to be material to patentability as defined in Title 37, Code of Federal Regulations, §1.56.

I hereby claim priority benefits under Title 35, United States Code, §119 (and §172 if this application is for a Design) of any application(s) for patent or inventor's certificate listed below and have also identified below any application for patent or inventor's certificate having a filing date before that of the application on which priority is claimed:

COUNTRY	APPLICATION NO.	DATE OF FILING	PRIORITY CLAIMED
Austria	A 647 /99	13/04/1999	yes

I hereby claim the benefit under Title 35, United States Code, §120 of any United States application(s) listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in the prior United States application in the manner provided by the first paragraph of Title 35, United States Code, §112, I acknowledge the duty to disclose information material to patentability as defined in Title 37, Code of Federal Regulations, §1.56 which occurred between the filing date of the prior application and the national or PCT international filing date of this application.

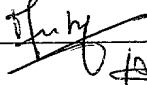

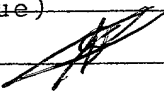
APPLICATION SERIAL NO.	U.S. FILING DATE	STATUS: PATENTED, PENDING, ABANDONED

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I further declare that all statements made herein of my own knowledge are true, and that all statements on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code, and that such willful false statements may jeopardize the validity of the application or any patent issuing thereon.

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3rd Inventor	(Alsters Paul)		Date	August 8, 2001
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The above application may be more particularly identified as follows:

U.S. Application Serial No.		Filing Date	
Applicant Reference Number		Atty Docket No.	
Title of Invention			



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And I hereby appoint John T. Miller, Reg. No. 21,120; Michael R. Davis, Reg. No. 25,134; Matthew M. Jacob, Reg. No. 25,154; Jeffrey Nolton, Reg. No. 25,408; Warren M. Cheek, Jr., Reg. No. 33,367; Nils E. Pedersen, Reg. No. 33,145 and Charles R. Watts, Reg. No. 33,142, who together constitute the firm of WENDEROTH, LIND & PONACK, L.L.P., attorneys to prosecute this application and to transact all business in the U.S. Patent and Trademark Office connected therewith.

I hereby authorize the U.S. attorneys named herein to accept and follow instructions from Patent Department DSM-Fine Chemicals Austria as to any action to be taken in the U.S. Patent and Trademark Office regarding this application without direct communication between the U.S. attorneys and myself. In the event of a change in the persons from whom instructions may be taken, the U.S. attorneys named herein will be so notified by me.

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